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Advanced Thermal Barrier System Bond Coatings for Use on Ni-, Co-, and Fe-Base Alloy Substrates

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ADVANCED THERMAL BARRIER SYSTEM BOND COATINGS FOR USE ON
Ni-, Co-, AND Fe-BASE ALLOY SUBSTRATES

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SUMMARY

Ni-, Co-, and Fe-base bond coatings containing between 16 to 38 wt % chromium, 6 to 20 wt % aluminum, 0.04 to 2.3 wt % ytterbium or 0.04 to 1.60 wt % yttrium were evaluated in a cyclic furnace between 1120 and 1175 °C. These bond coatings were plasma spray deposited in air onto Ni-, Co-, and Fe-base substrates and then plasma spray coated in air with 6.1Y₂O₃-or 8.0Y₂O₃-stabilized zirconia thermal barrier coating.

It was established that Ni-25Cr-6Al-Yb and Ni-35Cr-6Al-Yb bond coatings have optimum ytterbium concentrations at which maximum lives are obtained. In all three bond coatings, increasing chromium from about 16 to 38 wt % is very beneficial while the increase in aluminum from about 6 to 20 wt % is very detrimental. The M-16Cr-6Al-Yb(or Y), M-(25 to 35)Cr-(10 to 20)Al-Yb(or Y) bond coatings were not evaluated very extensively since these systems fail very rapidly as compared to M-(25 to 35)Cr-6Al bond coatings at optimum ytterbium concentrations (where M = Ni, Co, or Fe).

It was also found that by increasing the NiCrAlYb bond coating thickness from 0.010 to 0.020 cm, thermal barrier system life increased twofold.

Optimum ytterbium or yttrium concentrations and the thickness effect for Co-, and Fe-base bond coatings were not determined in this study. Also, not evaluated were the density effects of the three bond coatings studied, although their effects on the thermal barrier system lives are expected to be significant.

The weight gain (oxidation) data and the failures of these three bond coating systems are very similar. There appears to be no direct correlation between system to system lives and weight gains at failure. However, within a given system, failures of replicate specimens occurred at about the same weight gain, at approximately plus or minus 6 percent. The thermal barrier systems fail through the formation of a crack or cracks within the thermal barrier coating near the bond coating interface or/and at the substrate-bond coating interface. The locations of the failures depend upon the compositions of the bond coatings. It is estimated that the Ni-35Cr-6Al-1.1Yb/ZrO₂-6.1Y₂O₃ system is at least 40 percent better than the best Ni-35Cr-5.9Al-0.95Y/ZrO₂-6.1Y₂O₃ system reported in the literature. Furthermore, the experimental data show that the longest life was obtained with the FeCrAlYb/ZrO₂-Y₂O₃ system on the FeCrAlCo substrate followed by NiCrAlYb and CoCrAlYb bond coating thermal barrier systems on MAR-M-200+Hf and MAR-M-509 substrates, respectively.

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INTRODUCTION

Thermal barrier systems' lives have been mainly improved through optimization of yttrium, chromium, and aluminum concentrations in the bond coating, optimization of yttria concentration in the zirconia thermal barrier coating, and selection of the plasma spray deposition parameters. The original Ni-16Cr-6Al-0.6Y bond coating (ref. 1) was optimized to Ni-16Cr-6Al-0.15Y with about a 35 percent increase in thermal barrier system life at 1095 °C (ref. 2). Recently, NiCrAlY bond coatings have further been optimized for Cr, Al, and Y contents and the resultant Ni-35Cr-6Al-0.95Y bond coating thermal barrier system has been found to have a life four times longer at 1110 °C than the Ni-16Cr-6Al-0.15Y bond coating thermal barrier system (ref. 3). The data in Ref. 2 clearly shows that the best thermal barrier coating of those studied is $\text{ZrO}_2\text{-6.1Y}_2\text{O}_3$. The effects of plasma spray deposition parameters on thermal barrier system life have been reported in references 3 to 5. The improvement in thermal barrier system life has also been achieved by minimizing the compressive residual stresses in the thermal barrier coating, and also through segmentation and electron beam vapor deposition of the ceramic coating (refs. 6 and 7).

An attempt was also made to use zirconium instead of yttrium in the bond coating in order to improve thermal barrier system life (refs. 4 and 8). It was reported in reference 4 that the Ni-14Cr-14Al-0.1Zr bond coating was significantly better than the original Ni-16Cr-6Al-0.6Y bond coating. This author has not been able to find any additionally published data that show effects of other reactive metals than yttrium and zirconium in the bond coating on the thermal barrier system life.

It has been reported that the oxidation resistance of the bond coating has a very significant effect on thermal barrier system life (refs. 2 to 4, and 9 to 11). In reference 2 it was shown that increasing yttrium concentration in the bond coating decreased the thickness of the bond coating that was oxidized at the time the thermal barrier system failed. The data in reference 9 suggest that yttrium in the bond coating of the NiCrAlY/ $\text{ZrO}_2\text{-Y}_2\text{O}_3$ system seems to be diffusing toward the bond-thermal barrier coating interface. In reference 10 it was shown that the presence of yttrium in the bond coating shifts the failure of the thermal barrier system from the substrate-bond coating interface to within the thermal barrier coating near the bond coating interface. In reference 9 it was observed that oxygen diffusion and the formation of oxides in the bond coating occurs mostly along the boundaries of the plasma-spray particles. Thus, the rate of diffusion of a reactive metal atoms, dissolved in the NiCrAl matrix, toward the boundaries of the plasma sprayed particles might play an important role in determining the depth of oxygen penetration and oxide formation in the bond coating. To study the effect of a larger atomic radius reactive metal than yttrium in the bond coating on thermal barrier system life, ytterbium was chosen because its atomic radius is the largest (1.92 Å) of all the rare earth metals except europium (ref. 12). Thus, the diffusion of ytterbium atoms out of the plasma sprayed particles toward the boundaries of these particles could be quite different from that of the yttrium atoms. Furthermore, since the trivalent ionic radius of ytterbium is the smallest (0.858 Å) of all the rare earth metals except lutetium (ref. 13), it was believed that ytterbium ions may change significantly the morphology of the oxide or oxides formed along the plasma spray particles. As was already pointed out above, oxygen penetrates into the bond coating rapidly along the boundaries of these plasma sprayed particles in the bond coating. Yttrium atomic and ionic radii are 1.80 and 0.893 Å, respectively (refs. 12 and 13).

Consequently, this report presents data on bond coatings containing yttrium or yttrium reactive metal. The new NiCrAlYb, CoCrAlYb(or Y), and FeCrAlYb(or Y) bond coatings with 6.1Y₂O₃- or 8.0Y₂O₃-stabilized zirconia thermal barrier coating were evaluated in a cyclic furnace between 1120 and 1175 °C.

MATERIALS, APPARATUS, AND PROCEDURE

Chemical compositions of MCrAlYb, MCrAlY bond coatings (where M = Ni, Co, or Fe) and ZrO₂-Y₂O₃ or YSZ thermal barrier coatings, as well as all substrate materials used are reported in Table I. All spray powders had particle size distributions of 90 percent minimum of -200 mesh (74 µm diam) to 85 percent minimum of +325 mesh (44 µm diam). NiCrAlYb bond coatings were evaluated on conventionally cast MAR-M-200+Hf alloy substrate material while the CoCrAlYb and CoCrAlY bond coatings were evaluated on conventionally cast MAR-M-509 alloy material. The specimens of both of these substrates were nominally 2.5x1.2x0.25 cm and with all corners and edges rounded to about 0.25 cm diameter. The FeCrAlYb and FeCrAlY bond coatings were evaluated on Fe-22Cr-5.8Al-0.5Co substrate and the specimens were nominally 2.5x1.9x0.5 cm whose corners and edges were rounded to about 0.5 cm diameter.

Specimen surfaces were grit-blast cleaned with fresh high-purity alumina and the bond coating plasma spray deposited within 5 min. The YSZ coating was applied within 20 min of completing the bond coating deposition. All plasma spray depositions were done in open air. During plasma spray deposition, an attempt was made to maintain the plasma spray gun at 90° to the surface being sprayed but this was not always possible when spraying rounded surfaces. Further plasma spray deposition details are given in table II. To obtain greater coating thickness uniformity on all specimen surfaces, coating thickness on each surface was measured individually. This procedure allows to build up thin areas. The thicker thermal barrier coating on the last plasma spray deposited surface was brought into conformity with the thickness of the rest of the specimen through hand grinding on garnet abrasive paper. This approach was found to be a requirement to obtain acceptable test life reproducibility.

Specimens were tested in a cyclic furnace in air at test temperatures between 1120 and 1175 °C. The cycle in all cases consisted of a 9-min heat up, 60-min at temperature, and 60-min of cooling to about 300 °C. The temperature in the furnace, monitored with a Pt-Pt/13 percent Rh thermocouple, was estimated to be accurate to about ±10 °C at 1175 °C. Specimens were removed from the furnace between 350 and 400 °C after 1,2,3,4 and 12 cycles, and every 12 cycles thereafter for inspection. These specimens were cooled to room temperature and weighed. Because of their lower temperature, inspection cycles are more severe than the standard test cycles. Tests were continued until failure - the formation of a crack in the YSZ coating visible at the surface to the unaided eye.

RESULTS AND DISCUSSION

Nickel-based bond coatings. - The data in figures 1 and 2 are very similar to the NiCrAlY/ZrO₂-Y₂O₃ data reported in reference 3. The data in figures 1 and 2 show that the effect of yttrium concentrations in the bond coating on life passes through an optimum and that an increase of chromium in the bond

coating from 16 to 36 wt % is very beneficial, while an increase of aluminum in the bond coating from 6 to 19 wt % on thermal barrier system life is very detrimental for these air plasma sprayed bond coatings.

The effect of ytterbium on life passes through an optimum at about 1.1 wt %. This is higher in weight percent than the probable 0.8 wt % optimum yttrium concentration estimates for the Ni-35.0Cr-5.9Al-Y/ZrO₂-Y₂O₃ thermal barrier systems reported in reference 3. However, on an atomic percent basis, 1.1 wt % ytterbium and 0.8 wt % yttrium are about 0.34 and 0.47 at %, respectively. Thus, the lower optimum atomic percent ytterbium concentration has a more beneficial effect on thermal barrier system life than does the higher optimum atomic percent yttrium. The ytterbium bond coating system data cannot be directly compared with the yttrium bond coating system data because these two systems were studied at different temperatures. The NiCrAlYb/ZrO₂-Y₂O₃ systems were studied at 1125 and 1175 °C while the NiCrAlY/ZrO₂-Y₂O₃ systems were studied at 1110, 1160, and 1220 °C (ref. 3). A semiquantitative comparison of these two sets of data can be obtained by plotting "Ln of cycles to failure" versus "1/T K". Plotted data for selective NiCrAlYb/ZrO₂-Y₂O₃ and NiCrAlY/ZrO₂-Y₂O₃ systems are presented in figure 3. Assuming that NiCrAlYb/ZrO₂-Y₂O₃ and NiCrAlY/ZrO₂-Y₂O₃ behave similarly, straight lines were drawn through the NiCrAlYb/ZrO₂-Y₂O₃ points parallel to the least squares fit NiCrAlY/ZrO₂-Y₂O₃ lines. Using the data in figure 3, it is estimated that the Ni-36.1-1Cr-6.3Al-1.12Yb/ZrO₂-6.1Y₂O₃ system is at least 40 percent better than the Ni-35.0Cr-5.9Al-0.95Y/ZrO₂-6.1Y₂O₃ system. The superiority of the ytterbium over yttrium bond coating is also indicated by data at higher aluminum concentrations. The Ni-34.7Cr-18.7Al-0.92Yb/ZrO₂-8.0Y₂O₃ system at 1125 °C failed after about 200 1-hr cycles (fig. 1(b)), whereas the Ni-35.2±0.2Cr-18.5±0.2Al-0.80Y/ZrO₂-8.0Y₂O₃ system failed after about 60 1-hr cycles at 1110 °C (ref. 3). Consequently, 19 wt % aluminum bond coating containing ytterbium, although tested at higher temperature, is at least three times better than a similar yttrium doped bond coating system at somewhat lower temperature.

The effects of chromium and aluminum in the ytterbium-doped bond coatings on the thermal barrier system lives are very similar to those observed with the yttrium bond coatings. As the chromium concentration in the bond coating is increased from about 6 to about 35 wt % the relative change in life with ytterbium concentration increase is greater in the Ni-35Cr-6Al-Yb than in the Ni-16Cr-6Al-Yb bond coating systems (fig. 1(a)). Thus, for concentration increase from about 0.1 to 0.55 wt % in Ni-16Cr-6Al-Yb/ZrO₂-6.1Y₂O₃ and Ni-35Cr-6Al-Yb/ZrO₂-6.1Y₂O₃ systems, lives increased from about 380 to 480 and from about 500 to 1140 1-hr cycles, respectively.

The effect of an aluminum increase from about 6 to 19 wt % in the bond coating is very detrimental. Such an increase depresses the beneficial effect of ytterbium (fig. 1(b)). The lives of Ni-36.1Cr-6.3Al-1.12Yb/ZrO₂-6.1Y₂O₃ and Ni-34.7Cr-18.7Al-0.92Yb/ZrO₂-6.1Y₂O₃ systems are about 1350 and 260 1-hr cycles at 1125 °C, respectively. Furthermore, the change in life due to ytterbium increase from about 0.18 to 0.92 wt % in Ni-35Cr-19Al-Yb/ZrO₂-6.1Y₂O₃ system is only about 30 percent as compared to the about 100 percent estimated from figure 1 for the same ytterbium concentration increase in the Ni-35Cr-6Al-Yb/ZrO₂-6.1Y₂O₃ system.

The effect of bond-coating thickness on thermal barrier system life is shown in figure 4(a). As these data show, life increases at least twofold when the bond coating thickness is increased from 0.010 to 0.020 cm. From the data in Fig. 4(a) it is not possible to determine whether the bond coating thickness effect on life passes through an optimum. No data were obtained beyond 0.030 cm thickness since it is unlikely that bond coating thicknesses in excess of even 0.025 cm will be used. This limitation on bond coating thickness is imposed by the configurational limits of the hot components in an engine on which thermal barrier systems will be used.

The effect of thermal barrier coating thickness on life is shown in figure 4(b). These data show that an increase in thermal barrier coating thickness from about 0.028 to 0.080 cm decreases life by about one-half. The data in figure 4(b) are very similar to the data reported in reference 3 for the NiCrAlY/ZrO₂-Y₂O₃ systems.

The weight gain (oxidation) data for ytterbium bond coatings depend upon the compositions of these coatings. Selected representative weight gain data are presented in figure 5. From these data it may be concluded that there seems to be no direct universal relationship between the weight gain at failure and thermal barrier system life. However, replicates within each coating system show such a relationship. The "S" shaped curves in figure 5 are very similar to those reported for the yttrium containing bond coating systems (ref. 10). Thus, it is likely that both NiCrAlYb and NiCrAlY bond coatings are oxidized in a similar manner and their thermal barrier systems might also be failing in a similar manner. The similarities between the failure morphologies can be seen by comparing the photomicrographs in figures 6 and 7 with those reported in the literature for the yttrium-doped bond coating systems. The photomicrograph of the as-sprayed NiCrAlYb/ZrO₂-6.1Y₂O₃ thermal barrier system (Fig. 6(a)) is very similar to the NiCrAlY/ZrO₂-7.9Y₂O₃ thermal barrier system in reference 10. At failure, the bond coating in the NiCrAlYb/ZrO₂-6.1Y₂O₃ system is heavily oxidized and oxides are present along the splat boundaries and between the bond coating and zirconia. Normally the failure occurs first within the thermal barrier coating near the bond coating interface (figs. 6(b) and 7(a)). As these two photomicrographs show, at failure, depletion zones within the substrate near the substrate-bond coating interface have formed. This indicates a flux of aluminum to the bond coating. Indeed, this is exactly what has been encountered with NiCrAlY/ZrO₂-Y₂O₃ systems and reported in references 1, 7, and 10. The photomicrographs in figures 7(a) and (b) show that as the aluminum concentration in the bond coating increases, the failure location changes. All of the 25 or 35 wt % chromium bond coatings with nominally 6 wt % aluminum were found to fail within the ZrO₂-Y₂O₃ near the bond coating interface as shown in figure 7(a). However, the 25 or 35 wt % chromium bond coatings with nominally 11 or 18 wt % aluminum failed within the ZrO₃-Y₂O₃ near the bond coating interface and/or at the substrate-bond coating interface as shown in figure 7(b). At this point it is not possible to state where the crack first occurs with the 11 and 18 wt % aluminum bond coating systems because all samples were examined after the appearance of an external crack in the thermal barrier coating.

No attempt was made to determine the effect of bond and thermal barrier coating densities on the thermal barrier system life. However, since NiCrAlYb/ZrO₂-Y₂O₃ systems in many respects behave very similarly to the

NiCrAlY/ZrO₂-Y₂O₃ systems (ref. 3) it may be concluded that density effects on life will also be similar. Thus, the NiCrAlYb/ZrO₂-Y₂O₃ system should have optimum bond and thermal barrier coating densities.

Cobalt-based bond coatings. - The results from the evaluation of Co-16Cr-6Al-Y bond coatings with 6.1Y₂O₃-stabilized zirconia thermal barrier coatings at 1120 °C in a cyclic furnace showed that the bond coatings with 0.06 to 0.51 wt % yttrium are not very promising. This is suggested by the CoCrAlYb(or Y)/ZrO₂-6.1Y₂O₃ data in the lower part of figure 8. Very similar data were obtained for the CoCrAlYb(or Y)/ZrO₂-8.0Y₂O₃ systems but the lives in all cases were lower. By increasing chromium concentration from 16 to about 25 wt %, a nearly threefold life improvement was observed. Thus, Co-16.7Cr-5.9Al-0.13Y/ZrO₂-6.1Y₂O₃ and Co-25.1Cr-6.8Al-0.10Y/ZrO₂-6.1Y₂O₃ systems failed after about 58 and 160 1-hr cycles, respectively. This increase in life is very similar to that observed in NiCrAlY (ref. 3) and this suggests that higher chromium and yttrium concentrations in CoCrAlY might be beneficial. More extensive evaluation of CoCrAlY bond coatings having 25 to 35 wt % chromium, about 6 wt % aluminum, and 0.1 to 1.5 wt % yttrium were not carried out because the emphasis was shifted to CoCrAlYb coatings based on the results for NiCrAlYb bond coatings presented earlier in this report.

The data in figure 8 for CoCrAlYb/ZrO₂-Y₂O₃ systems were obtained only for low ytterbium concentrations. Higher ytterbium concentrations bond coatings were ordered but the vendor had difficulty in producing these powders. Only one yttrium and one ytterbium bond coatings in figure 8 can be compared. These data show that the Co-25.1Cr-6.8Al-0.10Y/ZrO₂-6.1Y₂O₃ system failed in about 160 1-hr cycles while the Co-27.8Cr-6.3Al-0.05Yb/ZrO₂-6.1Y₂O₃ system with lower reactive metal weight concentration failed in about 180 1-hr cycles. This is about a 10 percent increase in life. Note, that the atomic weight concentration of ytterbium is four times lower than the yttrium concentration, but that ytterbium is the more effective additive. This trend is very similar to that observed in the NiCrAlYb/ZrO₂-Y₂O₃ system. CoCrAlYb thermal barrier systems exhibited other trends that are similar to NiCrAlYb thermal barrier systems. At low ytterbium concentrations, the best CoCrAlYb bond coating seems to be the one containing 10 wt % aluminum (fig. 8). This is similar to the NiCrAlYb bond coating as shown in figure 1. Increasing the aluminum concentration to 20 wt % is very detrimental to thermal barrier system life (fig. 8 (top)).

The weight gain (oxidation) data for CoCrAlYb(or Y)/ZrO₂-Y₂O₃ thermal barrier systems are very similar to Ni-base bond coating systems in terms of the shapes of the weight gain versus time curves, and the lack of correlation between weight gain at failure and life for different compositions. However, the time required to obtain weight gains in the 10 to 15 mgs/cm² range at coating failure were less than by NiCrAlYb bond coating systems. It was found that 16 wt % chromium and 6 wt % aluminum bond coatings with low ytterbium or yttrium normally were almost completely oxidized at failure as shown in figure 9(a). The 25 and 35 wt % chromium with 6 wt % aluminum in the bond coating failed within the thermal barrier coating near the bond coating interface as shown by the photomicrograph in figure 6(b) for the NiCrAlYb/ZrO₂-Y₂O₃ system. It was observed that the 25 and 35 wt % chromium and about 11 wt % aluminum and low ytterbium or yttrium bond coatings with 6.1Y₂O₃- or 8.0Y₂O₃-stabilized zirconia coating also failed through the formation of cracks within the thermal barrier coating near the bond coating interface. The photomicrographs showed that these systems also had accumulation of oxides at the

substrate-bond coating interface but no apparent cracks. When aluminum concentration in the bond coating was increased to about 20 wt %, the thermal barrier system failed through the formation of cracks within the thermal barrier coating near the bond coating interface and/or at the substrate-bond coating interface (fig. 9(b)). Because the specimens were metallographically examined after failure occurred, it is not known which crack occurred first and caused the failure. The above results suggest that the ability of ytterbium to retard oxygen penetration into the bond coating decreases as the aluminum concentration in the bond coating increases from 6 to 20 wt %. This is very similar to the behavior observed with the NiCrAlYb/ZrO₂-Y₂O₃ system.

Based on the similarities between CoCrAlYb(or Y)/ZrO₂-Y₂O₃ and NiCrAlYb(or Y)/ZrO₂-Y₂O₃ systems, it may be assumed that the effects of bond coating thickness and density should also be similar, although the effects might be similar, the magnitudes of these effects remain to be determined.

Iron-based bond coatings. - The evaluations of the Fe-16Cr-6Al-Y/ZrO₂-8.0Y₂O₃ systems containing between 0.04 and 1.6 wt % yttrium showed that these systems are of very little use for they fail very rapidly (fig. 10). Increasing chromium concentration from 16 to 25 wt %, produced a dramatic increase in life. For example, Fe-16.5Cr-5.8Al-0.04Y and Fe-25.0Cr-6.5Al-0.05Y with ZrO₂-8.0Y₂O₃ failed after about 256 and 1255 1-hr cycles at 1125 °C, respectively. This increase is similar to that observed with NiCrAlY (ref. 3) and this suggests that higher chromium concentrations in FeCrAlY might be beneficial. The FeCrAlY bond coating was not further evaluated because of the results presented earlier for the benefits of ytterbium over yttrium in the bond coating. No optimization of the FeCrAlYb bond coating was done. The data in figure 10 were obtained for low ytterbium concentrations in the bond coatings. The data in figure 10 show that only one yttrium and one ytterbium bond coatings can be compared. The Fe-25.0Cr-6.5Al-0.04Y/ZrO₂-8.0Y₂O₃ system failed at 1125 °C after about 1255 1-hr cycles while the Fe-26.5 Cr-5.7Al-0.09Yb/ZrO₂-8.0Y₂O₃ system failed after 1550 1-hr cycles. This is an increase of about 23 percent. While the weight percent of ytterbium in the bond coating is about twice that of yttrium, however, on an atomic percent basis, the concentrations are about the same. The data in figure 10 also show that increasing ytterbium concentration is beneficial. Increasing ytterbium concentration from 0.09 to 0.2 wt % in the bond coating of the Fe-25Cr-6Al-Yb/ZrO₂-8.0Y₂O₃ system increased life from about 1550 to about 1740 1-hr cycles at 1125 °C. This corresponds to about 12 percent improvement. This suggests that further increases in ytterbium concentration of the bond coating might further improve the life of the thermal barrier system.

The data in figure 10 show that increasing the chromium concentration from the 25 to 35 wt % in the bond coating is beneficial. This increase corresponds to about 6 percent improvement in life. The data in figure 10 further show that increasing aluminum concentration in the bond coating from about 6 to about 18 wt % has a very detrimental effect on life. For example, Fe-37.4Cr-5.2Al-0.10Yb, Fe-37.8Cr-10.3Al-0.15Yb, and Fe-35.2Cr-18.3Al-0.12Yb with ZrO₂-8.0Y₂O₃ thermal barrier coating failed after 1760, 1204, and 203 1-hr cycles, respectively. This trend is very similar to those observed for the NiCrAlYb/ZrO₂-Y₂O₃ (fig. 1) and NiCrAlY/ZrO₂-Y₂O₃ systems reported in Ref. 3.

The weight gain data for FeCrAlYb(or Y)/ZrO₂-8.0Y₂O₃ systems in figure 11 are very similar to those of NiCrAlYb/ZrO₂-Y₂O₃ in figure 5. The failures of various FeCrAlYb(or Y)/ZrO₂-8.0Y₂O₃ systems were very similar to the failures

observed with Ni- and Co-base alloy bond coating systems. Because of these similarities, it may be concluded that the effects of the thicknesses and densities of both coatings in the FeCrAlYb(or Y)/ZrO₂-Y₂O₃ system are similar to those in the NiCrAlY/ZrO₂-Y₂O₃ system (ref. 3).

Comparison of nickel-, cobalt-, and iron-based bond coatings. - These coatings are not being compared at their respective optimum ytterbium or yttrium concentrations because Co- and Fe-based bond coatings were not optimized with respect to ytterbium or yttrium. Furthermore, Ni-, Co-, and Fe-based bond coating thermal barrier systems were only evaluated on MAR-M-200+Hf, MAR-M-509, and Fe-22Cr-5.8Al-0.5Co substrates, respectively.

From the data in figure 12 it is apparent that Fe-based bond coating thermal barrier system has the longest life. It is likely, that the life of this bond coating system may further be improved by increasing the ytterbium concentration as it is suggested by the data in figure 10. It is not possible to predict the magnitude of improvement because it is not known how far the ytterbium in Fe-35.4Cr-5.2Al-0.10Yb bond coating is from its optimum concentration. However, even if one was to assume that the 0.10Yb concentration is the optimum concentration, the Fe-35.4Cr-5.2Al-0.10Yb/ZrO₂-Y₂O₃ still has significantly longer life than the best NiCrAlYb/ZrO₂-Y₂O₃ system at its optimum ytterbium concentration reported in figure 1.

The data in figure 12 also suggest that Ni-based bond coatings are slightly better than the Co-based bond coatings. This is true for both, the NiCrAlYb and NiCrAlY bond coatings. It is unlikely that an increase of ytterbium concentration in Co-26.7Cr-5.6Al-Yb from 0.04 to 0.08Yb would improve this coating's life by about 5 times in order to have the same life as does the Ni-34.9Cr-6.9Al-0.08Yb bond coating (fig. 12). This observation is also applicable to Ni- and Co-based bond coatings containing yttrium (fig. 12, (bottom)). Consequently, it may be concluded, that within the limitations imposed in this study, Fe-based bond coating system has the longest life, followed by Ni- and Co-based alloy bond coating systems in that order.

SUMMARY OF RESULTS

Improved bond coatings for the two-layer thermal barrier systems to be used with Ni-, Co-, and Fe-base alloy substrates have been identified. The NiCrAlYb, CoCrAlYb(or Y), and FeCrAlYb(or Y) bond coatings with 6.1Y₂O₃- or 8.0 Y₂O₃-stabilized zirconia thermal barrier coating on Ni-, Co-, and Fe-base alloy substrates have been tested in a cyclic furnace between 1120 and 1175 °C. The evaluation of experimental data showed that:

1. The best NiCrAlYb bond coating was Ni-36.1Cr-6.6Al-1.12Yb and it is at least 40 percent better than the previously reported Ni-35.0Cr-5.9Al-0.95Y optimum bond coating. The data suggest that the optimum ytterbium concentration in the 25 and 35 weight percent chromium NiCrAlYb bond coatings is between 1.1 and 1.3 wt %.

2. CoCrAlYb bond coating is better than the CoCrAlY bond coating at low Yb and Y concentrations. Neither of these coatings were optimized. The best identified Co-base bond coating was Co-38.2Cr-10.3Al-0.07Yb.

3. FeCrAlYb bond coating is better than the FeCrAlY bond coating at low Yb and Y concentrations. Neither of these coatings were optimized with respect to Yb and Y. The best identified Fe-base bond coating was Fe-37.4Cr-5.2Al-0.10Yb.

4. The longest life was obtained with FeCrAlYb thermal barrier system on an iron-base substrate inspite of the fact that the bond coating was not optimized with respect to ytterbium followed by NiCrAlYb and CoCrAlYb bond coating thermal barrier systems on Ni-base and Co-base substrates, respectively.

5. Increases of chromium in all bond coatings from about 16 to 38 wt % are very beneficial while increases in aluminum from about 6 to 20 wt % are very detrimental for better quality bond coatings.

6. The data for NiCrAlYb bond coating thermal barrier systems show that an increase in the bond coating thickness from about 0.010 to 0.030 cm increases thermal barrier system life substantially. The increase in thermal barrier coating thickness from about 0.028 to 0.080 cm decreases thermal barrier system life significantly.

7. The weight gain (oxidation) data and the failure morphologies of the NiCrAlYb, CoCrAlYb(or Y), and FeCrAlYb(or Y) thermal barrier systems are very similar to those of NiCrAlY thermal barrier systems. The weight gain at failure depends primarily on the bond coating composition and on the processing parameters. Metallographically, failures were observed within the thermal barrier coating near the bond coating interface or both within the thermal barrier near the bond coating interface and at the substrate-bond coating interface depending upon the bond coating composition and in particular on aluminum concentration.

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TABLE I. - ACTUAL COMPOSITIONS OF SPRAY POWDERS AND SUPERALLOY SUBSTRATES^a

| Element | Composition, wt % | | | | | | | | |
|----------------|------------------------|------------------------|------------------------|------------------------|------------------------|---|--------------|-----------|----------|
| | NiCrAlYb | CoCrAlY | CoCrAlYb | FeCrAlY | FeCrAlYb | ZrO ₂ -Y ₂ O ₃ | MAR-M-200+Hf | MAR-M-509 | FeCrAlCo |
| Al | b _{5.0-19.0} | b _{5.0-7.0} | b _{5.5-21.0} | b _{5.0-7.0} | b _{5.0-19.0} | 0.001 | 5.21 | (c) | 5.8 |
| B | 0.003 | <0.001 | <0.001 | 0.004 | 0.001 | <0.001 | 0.02 | <0.001 | 0.04 |
| C | .007 | .012 | .010 | .011 | .014 | (c) | .17 | .65 | .10 |
| Ca | <.001 | <.001 | <.001 | <.001 | <.001 | .060 | (c) | .001 | .001 |
| Co | .030 | Major | Major | .035 | .038 | <.001 | 10.15 | Major | .51 |
| Cr | b ₁₇₋₃₇ | b ₁₆₋₂₅ | b ₂₅₋₃₈ | b ₁₅₋₂₅ | b ₂₆₋₃₈ | <.001 | 8.52 | 23.55 | 22.0 |
| Cu | .006 | .004 | .003 | .003 | .003 | (c) | .02 | <.01 | <.01 |
| Fe | .13 | .009 | .005 | Major | Major | .035 | .06 | .58 | Major |
| Hf | <.01 | <.01 | <.01 | <.01 | <.01 | 1.64 | 2.12 | .01 | <.01 |
| K | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 |
| Li | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 |
| Mg | <.001 | <.001 | <.001 | <.001 | <.001 | .022 | <.001 | <.001 | <.001 |
| Mn | .003 | .003 | .002 | .002 | .002 | .001 | <.01 | <.001 | <.001 |
| Mo | <.005 | .010 | .008 | <.005 | <.005 | <.005 | 5.81 | .005 | .005 |
| N ₂ | .016 | .018 | .022 | .030 | .028 | (c) | (c) | (c) | (c) |
| Na | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 | <.001 |
| Nb | <.01 | <.005 | <.005 | <.005 | <.005 | .005 | 1.19 | .005 | .005 |
| Ni | Major | .10 | .05 | .05 | .05 | .001 | Major | 10.62 | .09 |
| O ₂ | .016 | .021 | .018 | .018 | .016 | (c) | (c) | (c) | (c) |
| P | .004 | .006 | .006 | .004 | .005 | (c) | (c) | (c) | (c) |
| Pb | (c) | (c) | (c) | (c) | (c) | (c) | .005 | .005 | (c) |
| S | .003 | .002 | .003 | .003 | .003 | (c) | <.005 | <.005 | <.005 |
| Si | .028 | .024 | .021 | .025 | .015 | .055 | .15 | .22 | .08 |
| Sr | <.01 | <.01 | <.01 | <.01 | <.01 | <.01 | <.01 | <.01 | <.01 |
| Ta | <.05 | <.05 | <.05 | <.05 | <.05 | .02 | .06 | 3.77 | <.01 |
| Ti | <.005 | <.005 | <.005 | <.005 | <.005 | .035 | 2.06 | .16 | .01 |
| V | <.005 | <.005 | <.005 | .018 | .015 | <.001 | <.001 | .69 | .001 |
| W | <.01 | <.01 | <.01 | <.01 | <.01 | .01 | 11.62 | 6.96 | <.01 |
| Y | .005 | b _{0.06-0.51} | .005 | b _{0.04-1.60} | .005 | b _{4.8-6.3} | .001 | .001 | .001 |
| Yb | b _{0.08-2.33} | .001 | b _{0.05-0.16} | .001 | b _{0.09-0.20} | .001 | .005 | .005 | .005 |
| Zn | <.005 | <.005 | <.005 | <.005 | <.005 | <.005 | <.05 | <.05 | <.05 |
| Zr | <.005 | <.005 | <.005 | <.005 | <.005 | Major | .06 | .53 | .15 |

^aImpurity values reported in the table are the maximum values encountered.^bAluminum, chromium, yttrium, and ytterbium concentrations for various bond coatings and yttria in zirconia thermal barrier coatings used are given in tables presenting experimental data.^cNot determined.TABLE II - PLASMA SPRAY COATING DEPOSITION PARAMETERS^a

| Operating parameters | NiCrAlY | ZrO ₂ -Y ₂ O ₃ |
|---------------------------------------|-------------|---|
| Arc gas | Argon | Argon |
| Arc gas flow, 1/min | 26.0±0.5 | 28.3±0.5 |
| Powder gas | Argon | Argon |
| Powder gas flow, 1/min | 5.2±0.2 | 5.6±0.2 |
| Current, A | 400±5 | 600±5 |
| Voltage, V | 28±1 | 31±1 |
| Powder feed rate, gm/min | 12±1 | 15±6 |
| Stand-off-distance, cm | 12±2 | 12±2 |
| Bond coating thickness, cm | 0.020±0.002 | ----- |
| Thermal barrier coating thickness, cm | ----- | 0.039±0.003 |

^aStandard conditions used unless others are specified.

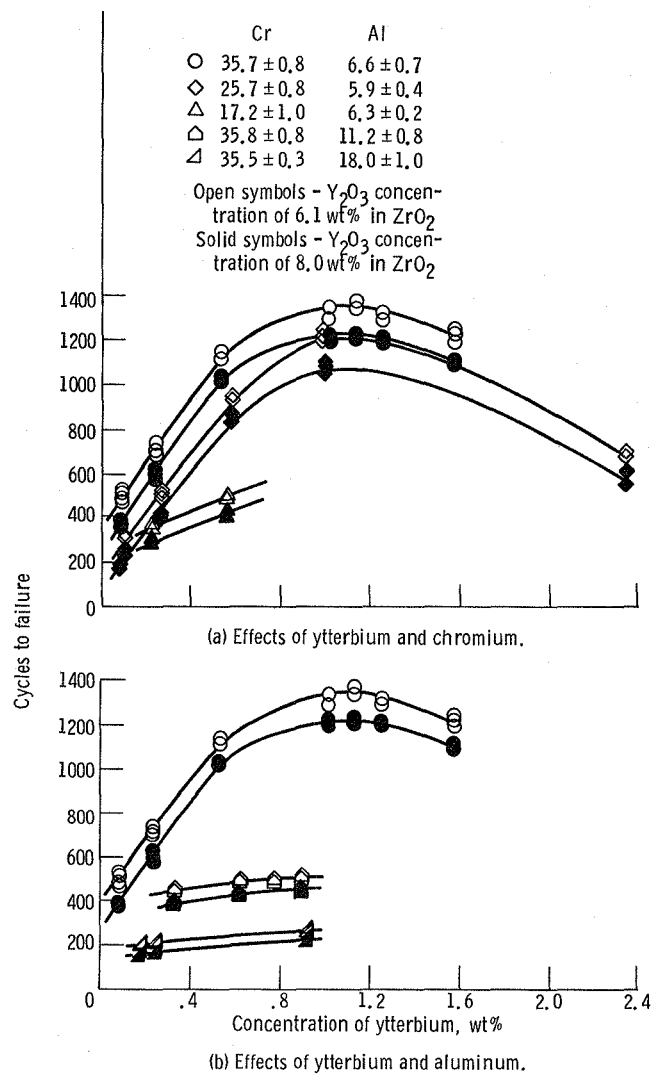


Fig. 1. - The effects of bond and thermal barrier coating compositions on the life of $NiCrAlYb/ZrO_2-Y_2O_3$ as determined in a cyclic furnace at $1125^\circ C$.

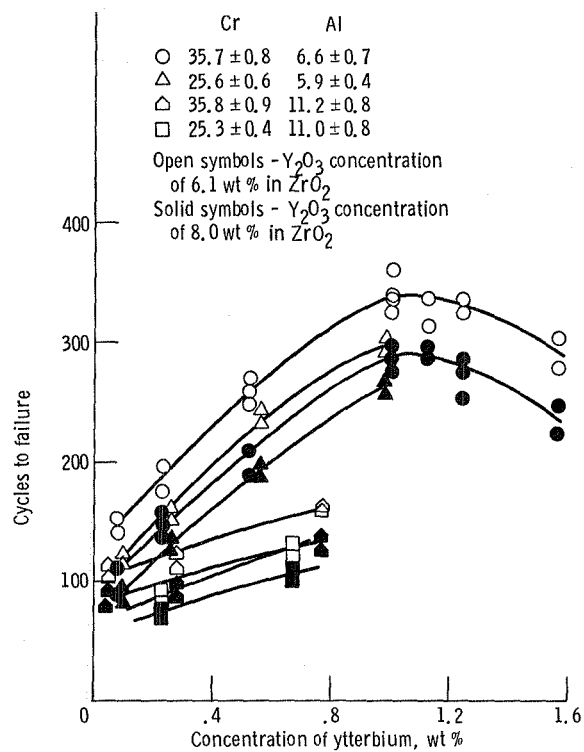


Fig. 2. - The effects of yttrium, chromium, and aluminum concentrations in the bond coating and yttria concentration in zirconia on the life of $\text{NiCrAlYb}/\text{ZrO}_2\text{-Y}_2\text{O}_3$ as determined in a cyclic furnace at 1175°C .

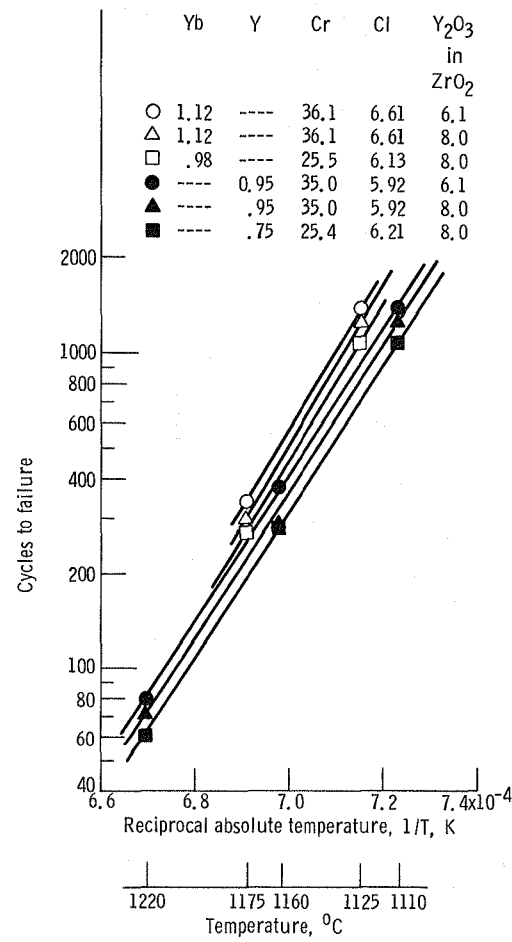
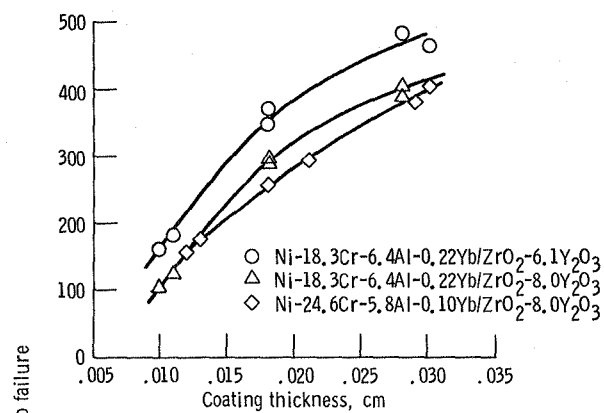
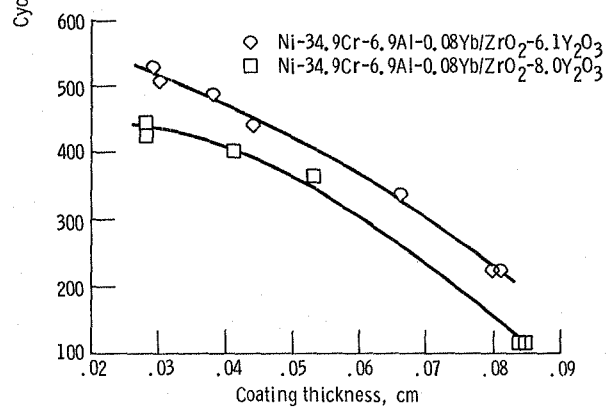


Fig. 3. - Comparison of lives of thermal barrier systems with NiCrAlYb and NiCrAl-Y bond coatings.



(a) Effect of NiCrAlYb bond coating thickness.



(b) Effect of ZrO₂-Y₂O₃ coating thickness.

Fig. 4. - The effects of coating thicknesses on the thermal barrier system life.

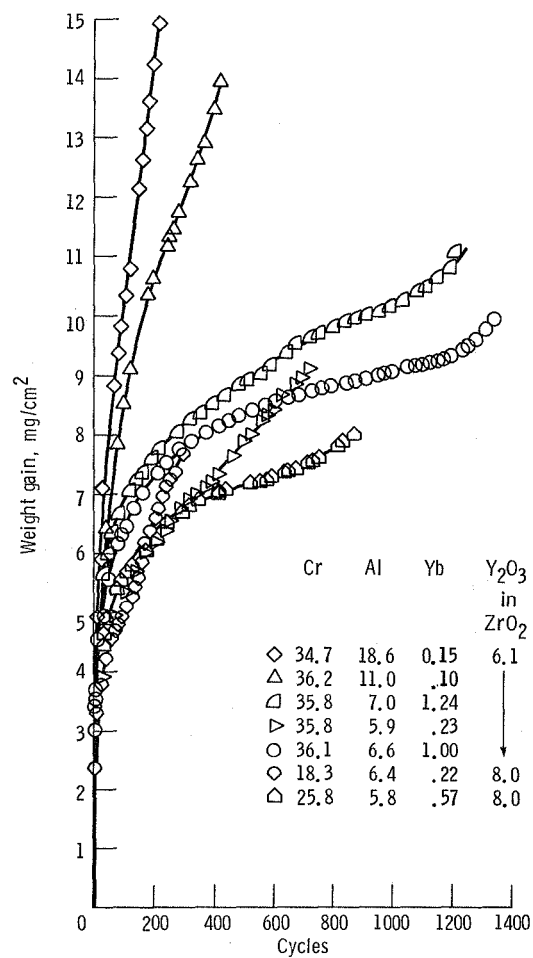
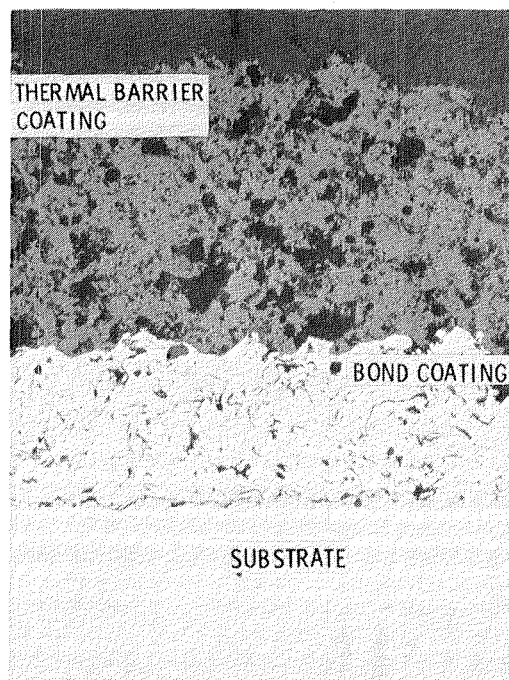
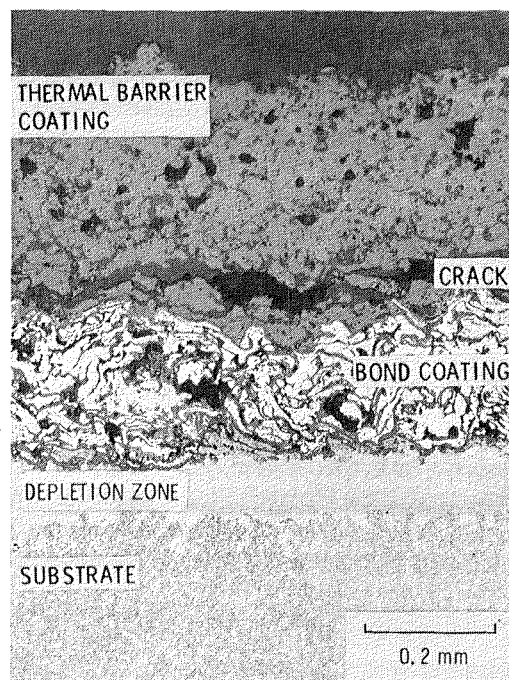


Fig. 5. - Oxidation behavior of NiCrAlYb/ZrO₂-Y₂O₃ thermal barrier systems up to visual failure of ZrO₂-Y₂O₃ as determined at 1125 °C.

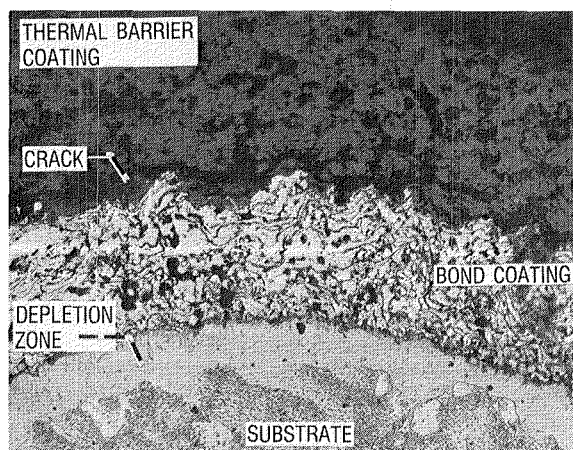


(a) As-spray coated only.

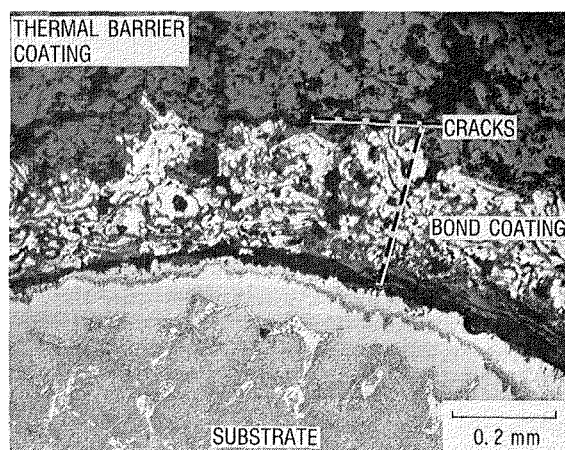


(b) After 482 1-hour furnace cycles to 1125°C.

Fig. 6. - Optical photomicrographs of NAR-M-200+ Hf specimens plasma spray coated with Ni-16.7 Cr-5.7 Al-0.55 Yb/ZrO₂-6.1 Y₂O₃ thermal barrier systems.



(a) Ni-25.8Cr-5.8Al-0.57Yb/ZrO₂-8.0Y₂O₃ after 186 1-hour cycles.



(b) Ni-36.0Cr-10.5Al-0.77Yb/ZrO₂-8.0Y₂O₃ after 162 1-hour cycles.

Fig. 7. - Optical photomicrographs of plasma spray coated MAR-M-200+Hf specimens that failed at 1175°C in a cyclic furnace.

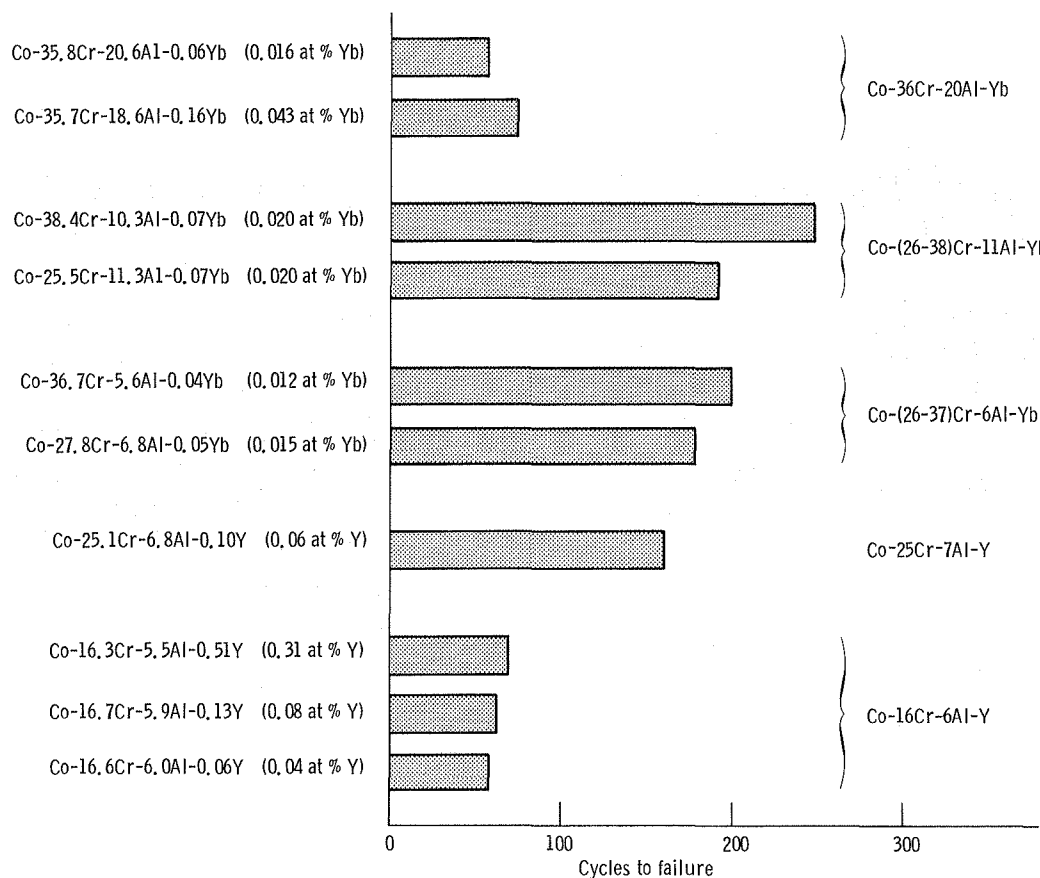
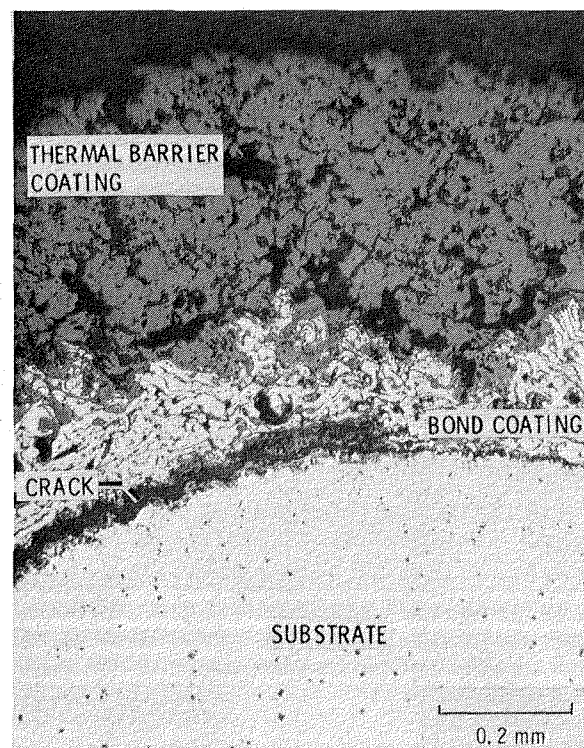


Fig. 8. - The effects of yttrium or yttrium, chromium, and aluminum concentrations in the bond coating on the CoCrAlYb (or Y)/ZrO₂-6.1Y₂O₃ thermal barrier system life as determined on MAR-M-509 substrate in a cyclic furnace at 1120 °C. (Each cycle to failure value is an average of at least three numbers)



(a) Co-16.6Cr-6.0Al-0.06Y/ZrO₂-6.1Y₂O₃ after 69 1-hour cycles.



(b) Co-35.8Cr-20.6Al-0.06Yb/ZrO₂-6.1Y₂O₃ after 56 1-hour cycles.

Fig. 9. - Optical photomicrographs of plasma spray coated MAR-M-509 specimens that failed at 1120 °C in a cyclic furnace.

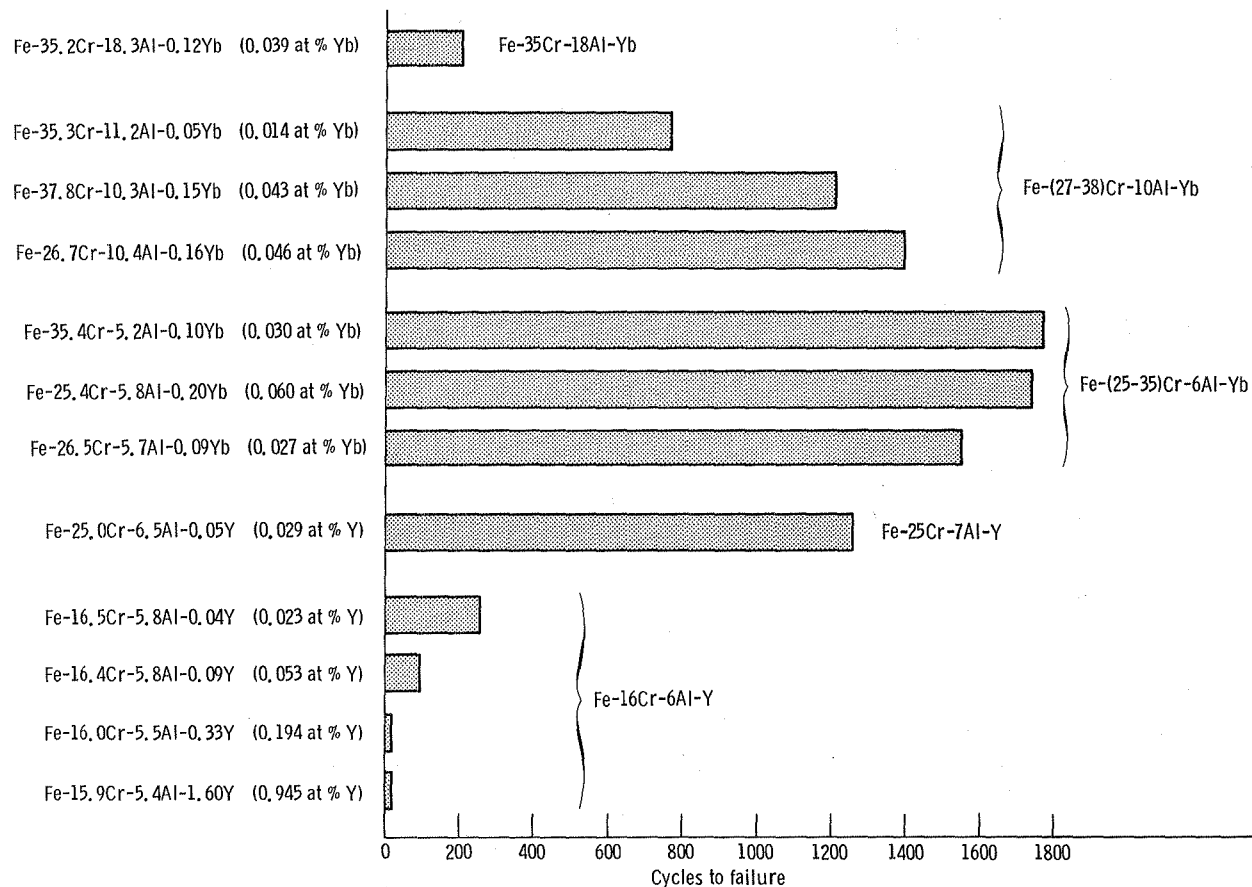


Fig. 10. - The effects of ytterbium or yttrium, chromium, and aluminum concentrations in the bond coating on FeCrAlYb (or Y)/ ZrO_2 -8.0Y $_2$ O $_3$ thermal barrier system life as determined on Fe-22Cr-5.8Al-0.5Co substrate in a cyclic furnace at 1125 °C. (Each cycle to failure value is an average of at least three numbers)

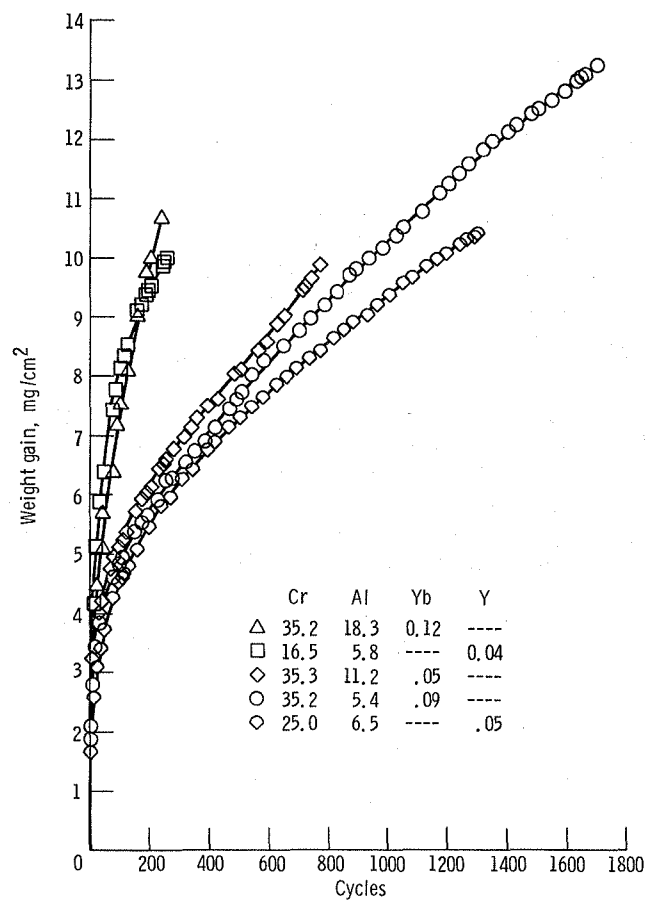


Fig. 11. - Oxidation behaviors of FeCrAlYb (or Y)/ZrO₂-8.0Y₂O₃ thermal barrier systems up to failures as determined in a cyclic furnace at 1125 °C.

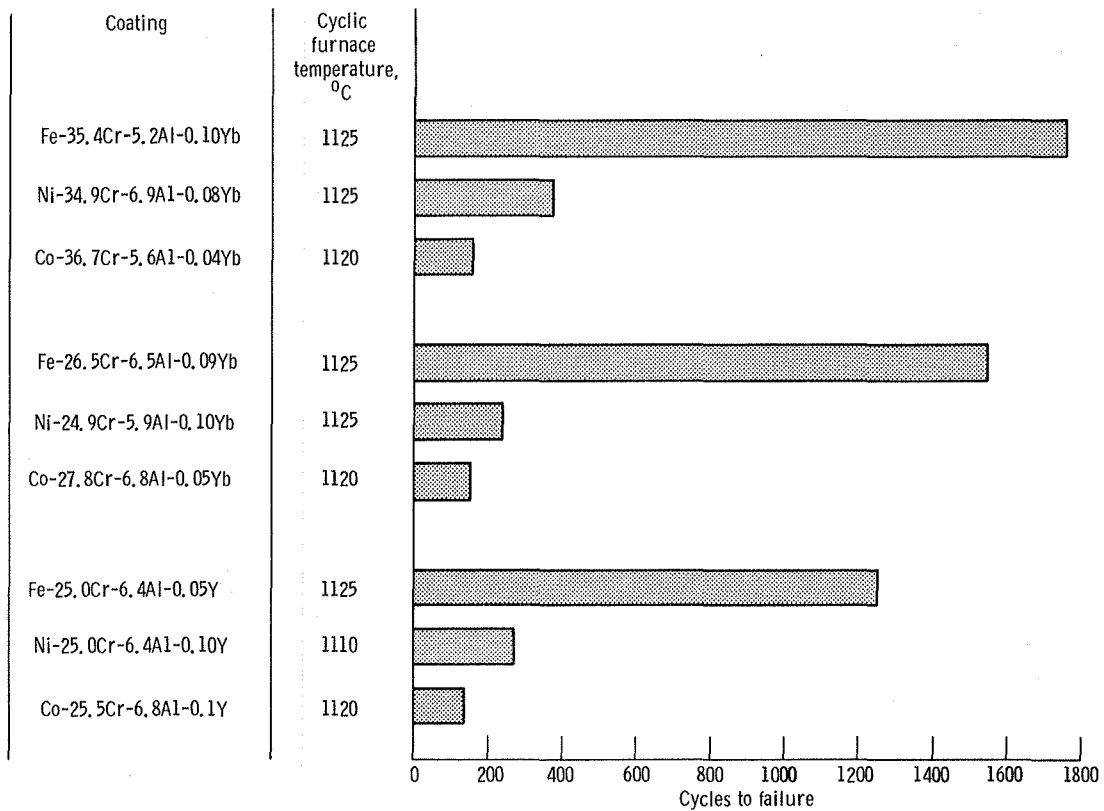


Fig. 12. - Comparison of Ni-, Co-, and Fe-base bond coating/ $\text{ZrO}_2\text{-}8.0\text{Y}_2\text{O}_3$ thermal barrier systems on MAR-M-200+Hf, MAR-M-509, and Fe-22Cr-5.8Al-0.5Co substrates, respectively.

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| 15. Supplementary Notes | | | | | |
| 16. Abstract New and improved Ni-, Co-, and Fe-base bond coatings have been identified for the $ZrO_2-Y_2O_3$ thermal barrier coatings to be used on Ni-, Co-, and Fe-base alloy substrates. These bond coatings were evaluated in a cyclic furnace between 1120 and 1175 °C. It was found that MCrAlYb (where M = Ni, Co, or Fe) bond coating thermal barrier systems have significantly longer lives than MCrAlY bond coating thermal barrier systems. The longest life was obtained with the FeCrAlYb thermal barrier system followed by NiCrAlYb and CoCrAlYb thermal barrier systems in that order. | | | | | |
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